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PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Takuya HIRAMATSU et al.

Group Art Unit: 1764

Serial No.: 09/524,575

Examiner: Hien Tran

Filed: March 13, 2000

For: HEATER AND CATALYTIC CONVERTER

**GROUP 1700** 

**DECLARATION UNDER 37 CFR 1.132** 

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Commissioner for Patents Washington, D.C. 20231

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Sir:

Akira TAKAHASHI, a citizen of Japan, residing at Parkcity
Ueda Riverview 1106, 102-1 Yoko-machi, Tenpaku-ku, Nagoya-city,
Aichi-prefecture, 468-0054 Japan
declares that:

- (1) he is a scientist who majored in the adsorption and catalyst technology for exhaust gas purification and he got his Ph.D. degree at the chemical engineering department, The University of Michigan, Ann Arbor, U.S.A.;
- (2) he is one of the inventors of U.S. Patent Nos. 5118644, 5229046, 5178809, 5691261, 5804521, 5884473, 6029441, 6051205, 6139808, 6171557, 6207604, 6294150, 6350416, 6500392, and 6517785;

- (3) he is familiar with the prosecution history of the aboveidentified application; and
- (4) he has compiled the following results to demonstrate that the features of the claims, particularly the features of claims 7-22 and 26, give properties or characteristics of an unexpected (and patentable) nature compared to related materials.
- (5) he has prepared the following comparative Table to demonstrate that the features of the claims, particularly the features of claims 7-22 and 26, give properties or characteristics of an unexpected (and patentable) nature compared to related materials.

## I. Stability Test

(1) Experiment and Results

A series of β-zeolite having the different SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios as is shown in Table 1 below, and a series of ZSM-5 having the different SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios as is shown in Table 1 below were used to demonstrate the stability for the retention of the specific area after they were exposed to 750°C or 850°C for 100 hours in "Fuel Cut Mode", so as to assess the stability of them when they are exposed to an exhaust gas.

After having been exposed to the predetermined temperature, the specific surface areas of the respective zeolite samples were determined by BET one-point method.

The results are shown in Table 1 below in terms of % against the specific surface area of the respective intact zeolite samples.

Table 1

				Retenti Area	on %	of	Specific	:	
Zeolite			β -zeo	ite			ZSM-5		
Sample No.	(1)	(2)	(3)	(4)	(5)	(1)	(2)	(3)	(4)
SiO2/Al2O3	25	95	110	210	290	50	150	200	250
Exposed 750	20	29	77	83	94	86	90	92	97
temperature 850	5	11	56	-80	86	71	77	87	81

## (2) Discussion

As is clear from the results shown in Table 1, β-zeolite clearly shows a preferable stability as adsorbent when whose SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio is 200 or more if β-zeolite is exposed to a temperature of 850°C with which a zeolite used as an adsorbent may encounter under actual driving conditions, even compared with ZSM-5 having a good thermal stability. On the other hand, the Document 94/11623 merely teaches the preference of SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio in the broader range on Page 4, Third Paragraph of the Document 94/11623.

Indeed, even in the case of  $\beta$ -zeolite whose SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio is 110, the stability is comparable to that of ZSM-5 under the exposed conditions of 750°C.

II. Comparison of Performance between Cross System vs. In-Line System. ľ

The Declarant compiled the following Comparative Table to compare the performance of the system disclosed in the Document with that of the present system.

In case of the Document WO 94/11623, the highest  $SiO_2/Al_2O_3$  of  $\beta$ -zeolite among those actually used for purification test is 180 in Examples 2 and 3. Thus, the results of those Examples are used in this Comparative Table.

In case of the present application, all data inclusive of Comparative Examples 1-7 are shown in Comparative Table below.

The reason is to show that  $SiO_2/Al_2O_3$  of 95 or less is not sufficient, and that the joint use of ZSM-5 with  $\beta$ -zeolite having  $SiO_2/Al_2O_3$  ratio of 210 can show some synergistic performance, compared with the joint use of ZSM-5 with that having  $SiO_2/Al_2O_3$  ratio of 25.

Comparative Table

		ا	Comparison of Performance	ormance		
	Constituti	Constitution of catalyst & adsorbent	adsorbent		Dorformana	
No. of	Zeolite	Amount	Type of	Total ami of	Emission in	
Example o	or (Si/Al	(g)pesn	System	Catalvet	ETO Toot	Furnication
Comparative				ac dimension	159	ratio
Framolo				loaded	(g/mile)	£
aldillav.				(g)		
Cxample 2	None	-	Cross	5.10	0 233	
	Beta	198	Ditto	5.10	0.161	900
	(180)				5	20.0
Example 3	None	1	Ditto	677	90,0	
	Reta	198	7450	100	0.106	
	(180)	2		7/0	0.065	62.4
Example 1	β (290)	250*	in-line	7.00	6,00	
Example 2	8(110)	252		26.1	0.045	78
Example 3	8 (910)	202	Diffe	9.39	0.059	72
	/0171d	1				
	WS7	220*	Difto	8.19	0.055	92
	(200)					3
Example 4	β(210)	253	Ditto	939	0.050	6
Example 5	$\beta$ (290)	250*	Citto	010	0.00	70
Example 6	A (210)	250		610	0.049	88
Evample 7	0/2/0	200	Unito	4.80	0.072	63
C. Translibio	0(230)	*DG7	Difto	21.47	0.042	80
C.variible o	6 (210)	210*	Ditto	23.72	0.039	82
Example 9	β (210)	210*	Oitto	21.93	0000	3
Comp. Exam.1	8 (95)	250*	Ditto	797	0.100	0 6
Comp. Exam.2	β (25)+				0.100	63
	ZSM	250*	Diffe	3 30		;
	(200)			999	0.098	65
Comp. Exam.3		250*	Ditto	7 0.9	0000	3,
Comp. Exam.4		250*	2++0	10.10	0.030	40
Comp. Exam 5	-	25.2	טייר	77.77	0.108	40
, L	1	207	UITTO	12.78	0.137	28
Comp. Cxam.o	1	253	Ditto	4.80	0.185	
Comp. Exam.	ZSIM (200)	253	Ditto	4.80	0.104	28
Remarks:						

Remarks:

(\*) mark denotes that a honeycomb having a through-hole structure were used in those systems.

(\*\*) mark denotes that the purification ratio is those obtained in the first cycle, while the rest is the purification ratio during the period of 0 to 150 seconds.

## (2) Discussion

In case of the present in-line type system,  $\beta$ -zeolite having  $\mathrm{SiO}_2/\mathrm{Al}_2\mathrm{O}_8$  ratio of 110 still shows a preferable performance, and is superior in the performance to that of  $\beta$ -zeolite in the cross system disclosed by WO 94/11623 both in reduction in emission and purification %.

This clearly establishes that the in-line system provided with  $\beta$ -zeolite having  $SiO_2/Al_2O_3$  ratio of 110 or more as an adsorbent shows a superior performance to that in the cross system provided zeolite having  $SiO_2/Al_2O_3$  ratio of 180.

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The undersigned further declares all the statements made herein of his own knowledge are true and that all the statements made on information and belief are to be true; and further that these statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States code and that such false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: April 16, 2003

AKITA TANANASHI